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Heavy metal pollution risk of desulfurized steel slag as a soil amendment in cycling use of solid wastes

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ABSTRACT

The by-product of wet flue gas desulfurization, desulfurized steel slag (DS), had chemical characteristics like natural gypsum that can be used to improve saline-sodic soil. However, contamination risk of heavy metals for cycling utilization of DS in agriculture was concerned mostly. Both pot and field experiments were conducted for evaluating the potential pollution risk of DS as the amendment of saline-sodic soil. Results showed that application of DS decreased the contents of Cd, Cu, Zn, and Pb, while significantly increasing chromium (Cr) content in DS-amended soils. The field experiment demonstrated that the migration of heavy metals (Cd, Zn, Cu, and Pb) in the soil profile was negligible. The application of DS at the dosage of 22.5–225 tons/ha significantly increased the Cr content in alfalfa (*Medicago sativa* L.) but lower than the national standard for feed in China (GB 13078-2017). DS altered the chemical fraction of heavy metals (Zn, Cu, and Pb), transferred exchangeable, reducible into oxidizable and residual forms in DS-amended soil. Application of DS combined with fulvic acid (FA) could effectively reduce the movement of heavy metals in soil and the accumulation of Cr in alfalfa. Based on our results, DS was a safe and feasible material for agricultural use and presented relatively little pollution risk of heavy metals. However, the results also showed that DS to a certain extent had a potential environmental risk of Cr if larger dosages of DS were used.

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Introduction

Steel slag is an industrial solid waste generated in the iron and steel production processes and its amount approximately accounts for 15%-20% of global steel and iron production. The disposal and storage of a large amount of steel slag not only occupy a lot of lands but also cause serious environmental problems (Fisher and Barron, 2019; Guo et al., 2018; Wang et al., 2021a; Zhang et al., 2020). Until now, steel slag mainly utilized in industrial fields, such as building or cement materials, carbon dioxide sequestration, and adsorbents for pollutants (Cha et al., 2006; Cui et al., 2021; Francisca and Glatsein, 2020; Kambole et al., 2017; Scott and Penn, 2021; Zhang et al., 2020). In agriculture, steel slag had been successfully used as producing fertilizer, liming material to neutralize acidic soils, and soil amendments to remedy heavy metal-contaminated soils (Annunziata et al., 2014; He et al., 2017; Matveeva et al., 2018; Wang and Cai, 2006; Wang et al., 2015; Wen et al., 2020).

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Fossil fuel combustion in the industry can produce large amounts of flue gas containing various air pollutants, such as sulfur dioxide (SO_2), which has caused a variety of air pollution problems. To meet clean air standards, almost every coal-fired power plant was asked to be equipped with a desulfurized facility to remove NOx and SO_2 in the flue gas (Chang et al., 2016). Among them, wet flue gas desulfurization (WFGD) was the most widely used technology for SO_2 removal because of its cheap, rich materials and the convenience of engineering applications (Cui et al., 2021; Koralegedara et al., 2019; Srivastava and Jozewicz, 2001). In the newly developed WFGD technology, industrial solid wastes, such as fly ash and steel slag, have been used as desulfurized absorbents (Yi et al., 2012; Yang et al., 2017). Despite the complex composition, steel slag is a good absorbent in desulfurization and denitrification. By using steel slag as desulfurizers and optimizing the operation conditions, the removal rates of SO_2 and NOx could reach 100% and 83.4%, respectively (Meng et al., 2018). Therefore, steel slag could replace lime as a new desulfurizer for wet flue gas desulfurization. Compared with other absorbents, steel slag as solid waste is almost free adsorbent to remove SO_2. Similarly, desulfurized steel slag (DS), a solid industrial by-product, was produced in the flue gas desulfurization process of steel slag.

The main component of desulfurized steel slag is calcium sulfate, which is similar to desulfurized gypsum. Steel slag is originally strong alkaline, but it becomes neutral and weak acidic substances after absorbing SO_2 in the process of flue gas desulfurization (Meng et al., 2018, 2019; Ying et al., 2021). Therefore, it could be expected that DS could be used as the amendment of saline-sodic soil. As a soil amendment, the use of desulfurized gypsum has been widely studied (Chen et al., 2001; Clark et al., 2001; Cui et al., 2021; Koralegedara et al., 2019; Zhao et al., 2019). It could alleviate the harms of high alkalinity and excessive salinity, and significantly improved the physical properties of soil. A meta-analysis compared 2658 pairs of data points (with and without desulfurized gypsum) in 59 regions of China (Wang et al., 2021b). It was found that the application of desulfurized gypsum could significantly increase crop yield by 91.2% ± 22.5% (average 95%), and reduce soil exchangeable sodium percentage (ESP) by 37.4% ± 9.6% and pH value by 8.1% ± 1.7%. We firstly conducted a field experiment using desulfurized steel slag to remedy saline-sodic soils in China (Ying et al., 2021). Our field experiment showed the application of DS could prominently reduce soil pH, salinity, and alkalinity, and improve soil physical properties.

As the steel slag has great application potential in flue gas desulfurization, the wet desulphurization using steel slag has been operated stably in industrial-scale devices. This technology not only saved the cost of desulfurization but also fully utilized the solid waste. At the same time, the by-product of flue gas desulfurization could provide abundant and low-cost materials as a potential soil amendment, which is an example of the beneficial recycling utilization of solid wastes. However, steel slag contains some dangerous heavy metals, such as chromium (Cr) (Navarro et al., 2010; Piatak et al., 2019; Proctor et al., 2000; Wang et al., 2021a; Zhang et al., 2020). The risk of heavy metal pollution caused by the agricultural application of steel slag are worried (Mäkelä et al., 2012; Pistocchi et al., 2017). Wang et al. (2022) reported the concentration, form, and mobility of heavy metals in desulfurized sludge from coal-fired power plants. Their results showed that desulfurized sludge used for landfills might cause harmful pollution of Cd, Hg, and Zn. Many previous studies indicated that desulfurized gypsum as the soil amendment was safe for soil and plant system, which was considered as a promising material to remedy saline-sodic soils (Chen et al., 2015; Liu et al., 2021; Wang et al., 2017). Although the use of steel slag for agricultural purposes is not new, to our knowledge, there is no available information on the potential pollution risks of heavy metals in DS as soil amendment. Based on our successful application of DS in improving the saline-sodic soils, it is worth studying the pollution risks of heavy metals in DS as the soil amendment. The objectives of our study were to evaluate the heavy metal risk and application potential of solid waste DS as the amendment of saline-sodic soils and to provide a practice possibility for win-win-win strategy, flue gas desulfurization, soil improvement, and utilization of solid wastes.

1. Materials and methods

1.1. Experimental materials

The flue gas desulfurized steel slag (DS) used in this study was collected from the Baotou Iron and Steel Group, Inner Mongolia, China. The DS was ground into powder to pass through a 100-mesh sieve for material characterization. The main chemical composition and minerals of DS were characterized by X-ray fluorescence (XRF) spectroscopy (Bruker S8 TIGER, Germany) and X-ray diffraction (XRD) spectroscopy (Bruker D8 Advance, Germany), respectively. The contents of heavy metals (Zn, Cu, Cd, Pb, Cr) in DS were determined using an inductively coupled plasma-mass spectroscopy (ICP-MS, PerkinElmer NexIONTM 300X, Perkinelmer, USA). The scanning electron microscope coupled with X-ray energy-dispersive spectrometer (SEM-EDS, ZEISS GeminiSEM 300, Carl Zeiss AG, Germany) analyses were performed to obtain the surface structure, and surface elemental composition of the DS.

The field experiment was performed on a typical saline-sodic soil located in the middle reaches of the Yellow River, Inner Mongolia, China (107°34′53″E, 40°58′47″N). Soil particle composition is shown as grit (<2 mm and >0.05 mm): 15.72%, silt content (<0.05 mm and >0.002 mm): 48.84%, and clay content (<0.002 mm): 35.44% by the pipette method. According to the WRB classification, the soil of the test site is classified as solonetz. The soil properties were as follows: pH 9.1 (1:2.5 *m*/*V* soil/water), electrical conductivity 4.29 dS/m, exchangeable sodium percentage (ESP) 42.5%, sodium adsorption ration 12.49, and cation exchange capacity (CEC) 13.8 cmol(+)/kg. The key physical and chemical properties of soil were determined by the routine procedures (Lu, 2000). Fulvic acid (FA) was purchased from an accredited manufacturer.

1.2. Experiment design

The pot experiment included eight treatments with three replicates in a completely randomized design. The soil used for the pot experiment was sampled from the field

experimental site in June 2019. The treatment included the control (CK, without DS), DS amendment (1%, 5%, and 10%), and fulvic acid (FA) alone (1%), and DS (1%, 5%, and 10%) + FA (1%), respectively. The application rate of DS was determined by comprehensively considering the preliminary results of field experiment and the dosage of desulfurization gypsum reported in the literature (Liu et al., 2021; Zhao et al., 2019). Based on the type of amendment and the dosage of DS, the treatments were named as the CK, DS1, DS5, DS10, DS1+FA, DS5+FA, and DS10+FA, respectively. The DS was ground to pass through a 2 mm sieve before being applied to the soil. The DS amendments were mixed with the soils at the required rate. The mixture of soil and DS in each pot was 5 kg. It is estimated that the dosage of DS is equivalent to the addition of 22.5, 112.5, and 225 tons/ha (the weight of a 20 cm soil layer per ha was 2.25×10^6 kg). The mixture of soil and DS was maintained at the saturated water capacity and incubated in the greenhouse. After incubation for 30 days, 20 alfalfa (*Medicago sativa* L.) seeds in each pot were sown on 25 April 2020. During the incubation period, soil water content was maintained at the equivalent of 70% of field capacity by weekly adjustment based on weight. Alfalfa was harvested two times in 2020 and one time in 2021. The above-ground biomass of alfalfa was harvested, dried at 70°C, and weighed. The dry weight of alfalfa from the three times harvest was summed and presented as cumulative dry biomass weight. After alfalfa harvesting, soil samples were collected using a soil sampler. The soil from three points in each pot was randomly taken and homogenized to form a composite sample for each treatment. The soil samples were air-dried and then ground, and then the plant roots were manually picked out, sieved through 2 mm, and then stored in self-sealed bags for further testing.

The field experiment, covering a total area of 10 ha, was performed in Urad Front Banner, Bayannur City, Inner Mongolia, China. The experimental field was separated into three plots of 3.33 ha for each plot. In 2015, 2016, and 2018, DS was applied to the soil surface at the content of 180 tons/ha. Then the soil is ploughed to a depth of about 20 cm and leveled. Alfalfa (*Medicago sativa* L.) was planted in each plot and harvested three times a year. In June 2019, soil samples of 1, 3, and 4 years after DS application were collected and named as T1, T3, and T4, respectively. The saline-sodic soil without DS was sampled as a blank control group (CK). In each treatment plot, three similar areas are divided, representing three repetitions of the same treatment, and sampled. The soil profile of each treatment plot is excavated to a depth of 100 cm and mined into four parts according to the layers determined onsite, which are 0-10, 10-26, 26-48, and 48–85 cm respectively. In each treatment plot, 1 square meter of alfalfa is randomly selected, trimmed, washed with deionized water three times, dried at 70 °C, and then ground through a 2 mm sieve. A detailed description of the field experiment was presented in our previous paper (Ying et al., 2021).

1.3. *Total and extractable metals analyses*

The dried plant material was digested with HNO_3-H_2O_2 (7:1, *V/V*) in the microwave oven (MASTER 40, SINEO, China), and the concentrations of heavy metals (Zn, Cu, Cd, Pb, Cr) were analyzed using ICP-MS. The total concentrations of heavy metals in soil were analyzed by ICP-MS after microwave digestion with HNO_3-HF-H_2O_2 (6:2:2, *V/V/V*). In order to check the analysis quality, reagent blanks, replicate samples, and standard reference materials GBW07405 (GSS-5) were measured. The enrichment factor (EF) of heavy metals was calculated as the content of heavy metals in alfalfa divided by the total content of heavy metals in corresponding soil.

Bioavailable heavy metal was extracted by shaking the centrifugal tube with 2 g soil at a 1:10 (soil: water) ratio (*m/V*) at 250 r/min for 2 hr with 0.1 mol/L $CaCl_2$ and 0.05 mol/L EDTA solution, respectively. After centrifugation and filtration, concentrations of heavy metals in the extract solution were determined by ICP-MS. All filtrates were filtered with a 0.45 μm membrane filter before determination. The extraction percentage of heavy metals was calculated as the contents of $CaCl_2$ or EDTA-extractable metals divided by the total content of heavy metals in soil.

1.4. *Chemical forms of heavy metals in soils*

The fractionation of heavy metals in the soil was determined using the BCR sequential extraction procedure proposed by the European Community Bureau of Reference (Fuentes et al., 2004; Nurmesniemi et al., 2010; Smeda and Zyrcniki, 2002). The BCR extraction procedure has been widely used to evaluate the speciation of heavy metals in various substrates, such as soil and industrial waste. Briefly, a soil sample of 0.159 mm was weighed and placed in a 50 mL polyethene centrifuge tube, and then a four-step sequential extraction procedure was carried out. The exchangeable (F1), Fe-Mn oxides-bound or easily reducible (F2), organically bound fraction or oxidizable (F3) and residual fractions (F4) were sequential extracted using 0.1 mol/L CH_3COOH, 0.5 mol/L NH_2OH•HCl, 0.02 mol/L HNO_3 in 30% H_2O_2 and 3.2 mol/L NH_4OAc in 20% HNO_3 (pH 2.0), and HNO_3-HF-HCl solutions, respectively. The concentration of heavy metals from four form extractants was determined with ICP-MS after the filtrate was filtered by a 0.45 μm membrane filter.

1.5. *Data analysis*

Data analysis was performed using Excel 2019, SPSS software package (version 26.0) (SPSS Inc., Chicago, USA), and R 4.1.1. All results were expressed as the arithmetic mean value ± standard deviation. All figures were drawn by Excel 2019, Adobe Illustrator CC2018, Origin Pro 8.0, and R 4.1.1. The correlation coefficient was conducted using SPSS version 26.0. A one-way analysis of variance (ANOVA) was used for statistical analysis, followed by the Duncan test to determine the significant difference ($p < 0.05$ and $p < 0.01$) between the means of different treatments.

2. Results

2.1. *Characterization of desulfurized steel slag*

The DS was found to be rich in calcium (16.8%), sulfur (11.5%), iron (9.1%), silicon (4.4%), aluminum (2.1%), and magnesium (1.1%) elements (Appendix A Table S1), which mainly was

Table 1 – Heavy metal content (mg/kg) of DS-treated soils and related national standards of China.

Treatment		Cr	Cu	Zn	Cd	Pb
CK		66.7 ± 2.29cC[a]	41.91 ± 1.13aA	77.23 ± 2.69aA	0.13 ± 0.01aA	18.79 ± 0.24aA
Pot experiment	DS1	80.66 ± 3.82bc	40.57 ± 2.43a	76.1 ± 1.98ab	0.11 ± 0.02ab	18.47 ± 0.76ab
	DS5	102.89 ± 6.84b	35.41 ± 0.93ab	68.6 ± 3.28abc	0.11 ± 0.01ab	16.87 ± 0.84abc
	DS10	151.27 ± 11.44a	25.84 ± 7.48b	54.62 ± 9.87d	0.06 ± 0.02cd	12.57 ± 3.67c
	FA	62.49 ± 2.66c	36.82 ± 3.99ab	63.54 ± 2.5cd	0.09 ± 0bc	16.51 ± 0.08abc
	DS1+FA	77.53 ± 7.78bc	39.56 ± 3.12a	64.36 ± 1.7bcd	0.1 ± 0.01b	17.22 ± 0.65ab
	DS5+FA	98.94 ± 12.93b	33.48 ± 7.64ab	57.33 ± 8.45cd	0.05 ± 0.02d	15.81 ± 2.75abc
	DS10+FA	148.66 ± 2.15a	26.99 ± 5.88b	54.06 ± 6.38d	0.04 ± 0.02d	13.91 ± 2.65bc
Field experiment	T1	140.89 ± 17.28A	35.29 ± 0.51B	59.83 ± 2.84B	0.04 ± 0.01B	17.02 ± 1.4B
	T3	123.58 ± 1.85B	35.45 ± 4.01B	59.5 ± 11.39B	0.05 ± 0.02B	15.77 ± 0.65B
	T4	127.11 ± 10.93B	30.29 ± 1.45B	61.82 ± 2.94B	0.03 ± 0.01B	16.98 ± 0.92B
GB 15618-2018	pH > 7.5	250	190	300	0.6	170
	6.5 < pH ≤ 7.5	200	100	250	0.3	120
GB 4284-2018	A	500	500	1200	3	300
	B	1000	1500	3000	15	1000
EUL (1986)		1000	1000	2500	20	750
F value	DS	102.08**[b]	6.937**	7.673**	15.49**	4.85*
	FA	0.89	0.52	11.544**	23.53**	0.709
	DA×FA	0.01	0.30	1.17	1.93	0.63

FA: fulvic acid. EUL: European Union limits for sludge as fertilizer.

[a] Different letters indicate significant differences in heavy metal contents in soil between different treatments. Capital and small letters represent field and pot experiment, respectively.

[b] The * and ** indicate the significant difference of 5% ($p < 0.05$) and 1% ($p < 0.01$), respectively.

present as $CaSO_4 \cdot 2H_2O$ (Appendix A Fig. S1). The content of heavy metals in DS was very low and below the Control Standards of Pollutants in Sludge for Agricultural Use (GB 4284-2018), except for Cr (Table 1). The content of all heavy metals in DS was also lower than the sludge application standard allowed by European legislation (86/278/EEC, 1986). The BCR sequential extraction results of DS were included in Appendix A Table S2, indicating that the exchangeable or soluble form of Cr in DS was negligible. The reducible and oxidizable fractions of Cr accounted for 14% and 5% of the total Cr. The largest fractions were found to be residual form, which accounted for 81% of the total Cr, indicating that the bonding mechanism of Cr in DS is very stable and its mobility is poor. Therefore, the application of DS itself to agricultural soils can be considered safe. One concern when using DS as a soil amendment is the potential toxicity of the Cr to plants.

The morphology of DS particles was shown in Fig. 1. The SEM image revealed the heterogeneity of DS that was made up of particles or aggregates of different nature and sizes. The size of DS particles is mainly 1-50 μm and has a wrinkled surface with some cracks (Fig. 1a). The DS particles separated from the DS-treated soils exhibited mostly a plate shape, and the surface became rough (Fig. 1b). The surface of DS was observed to be covered with many fine particles. After applying fulvic acid, the surface of DS particles was further roughened and became loose particles (Fig. 1c). EDS results confirmed that Cr was evenly distributed on the surface of particles (Fig. 1d). The distribution of elemental sulfur and calcium exhibited basically the same distribution pattern, but elemental sulfur was more concentrated on the rough parts of the particle surface. It is expected that the rough surface of steel slag is easier to absorb sulfur dioxide, and SO_2 is easily adsorbed on the surface of small particles and in the pores of large particles. XRD results further supported the existence of $CaSO_4 \bullet 2H_2O$ in DS (Appendix A Fig. S1).

2.2. *Heavy metal content in DS-treated soils*

The content of heavy metals in the soil of the pot experiment was shown in Table 1. The content of all heavy metals in the DS-amended soils was far below the risk control standard for soil contamination of agricultural land in China (GB 15618-2018) (MEE and SAMR, 2018). The content of Cr in the DS-treated soils increased with the increased amount of DS application. Compared with the soil without DS application (CK treatment), the DS5 and DS10 treatments significantly increased the content of Cr in soils ($p<0.05$). The contents of Cd, Cu, Pb, and Zn in DS-treated soils slightly decreased relative to the control soil. A significant decrease was found in the DS10 treatment. Application of DS combined with FA had lower content of Cr soils than the DS alone treatment. It could be concluded that when the application amount is reasonable, DS as a soil conditioner is safe concerning heavy metals. Two-way ANOVA of DS and FA alone and combined application (Table 1) showed that the contents of Zn and Cd in soils were significantly influenced by each of the two factors separately. However, their interaction was not significant statistically.

The field experiment indicated that DS application to the soil resulted in the significant accumulation of Cr ($p < 0.05$) in the topsoil layer (0-10 cm), which decreased with increasing the application year. The content of Cr in soil increased from an average of 66.7 mg/kg soil in the CK to 140.9, 123.6, and 127.1 mg/kg soil in T1, T3, and T4 treatments, respectively. The average initial Cr content in our soil (Table 1) was lower than the background level found in Chinese soils which is 90 mg/kg (GB 15618-1995). Similar to the pot experiment, the content of

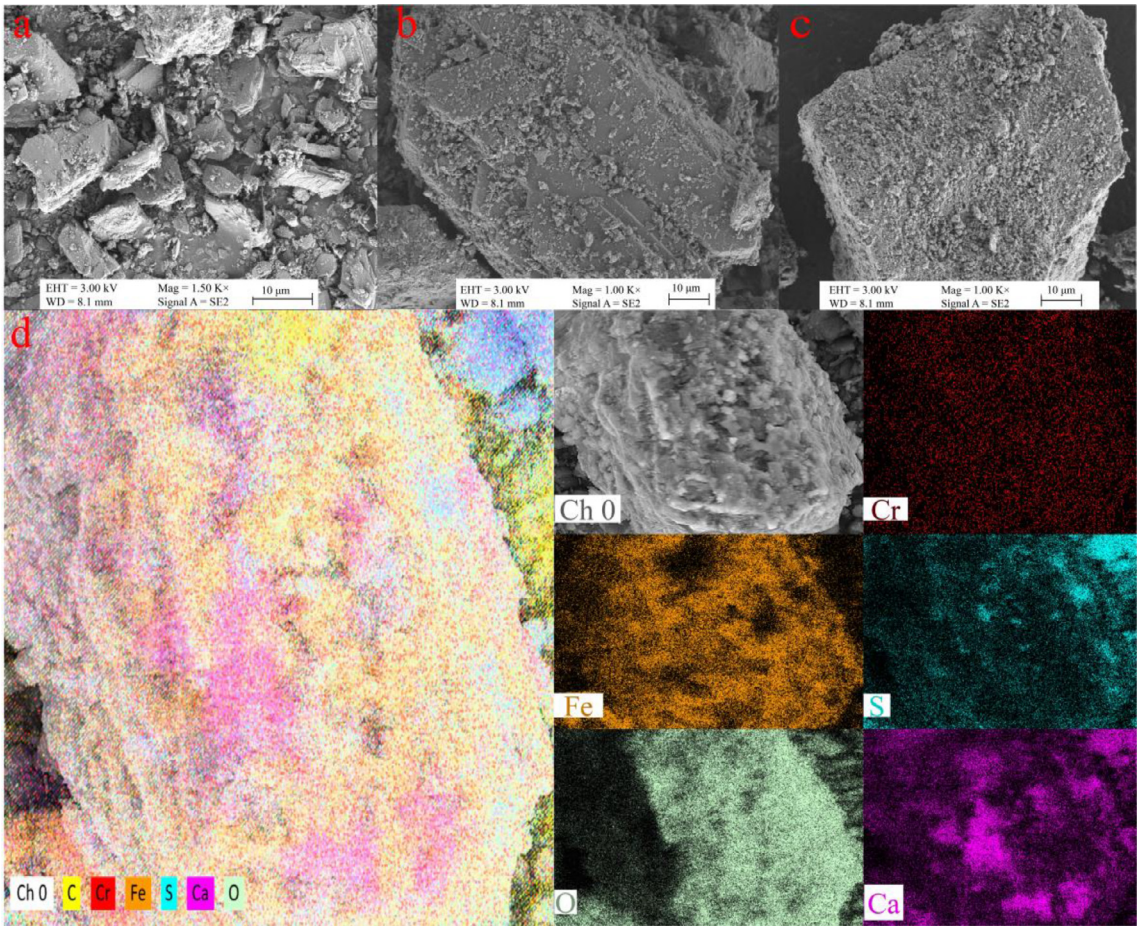

Fig. 1 – Surface morphology images and elemental distribution maps of desulfurization steel slag (DS) obtained by SEM/EDS. (a) DS particles; (b) DS particles isolated from DS10 treatment; (c) DS particles isolated from DS10+FA treatment; (d) EDS elemental maps for Cr, Fe, S, O, and Ca; 'Ch 0' represents SEM image corresponding to EDS area.

Cr in the field experimental soils was below the risk control standard for soil contamination of agricultural land in China (GB 15618-2018). The recovery of Cr in DS-treated soil was calculated and found to be around 100% in T1, confirming that most of the added Cr remained in the topsoil. The addition of Cr with the DS was stable in the soil. The variation of heavy metal contents in the soil profile was shown in Fig. 2. The application of DS significantly reduced the content of heavy metals, Cd, Cu, Pb, and Zn in the soil layer of 0-10 cm. Although the topsoil (0-10 cm) of DS-treated soil had significantly increased content of Cr, no significant difference was observed on the soil layers of 26–48 and 48–85 cm, indicating that mobility of Cr in the soil profile was negligible within a few years.

2.3. *Accumulation of heavy metals in alfalfa*

Due to the high salinity and alkalinity of original saline-sodic soil used in the pot and field experiment, the alfalfa could not germinate. No biomass of alfalfa could be obtained in the control treatment soil. The application of DS and fulvic acid alone or combined significantly enhanced the biomass of alfalfa compared with the control treatment (Appendix A Fig. S2). The combined application of DS with FA had a higher alfalfa yield than DS alone, and DS10+FA treatment had the largest increase in alfalfa biomass. DS addition alone or combined with FA evidently increased Cr accumulation in the alfalfa and the largest accumulation of Cr was found in the DS alone application (Fig. 3b). Compared with the DS alone treatment, the DS+FA treatments decreased the content of Cr in alfalfa and significant differences were achieved in the second and third harvest. Regardless of the dosage of DS application, the content of Cr in alfalfa grown in the DS-treated soils was lower less than the national hygienic standard for feed (GB 13078-2017), which was 5 mg/kg for Cr.

In the field experiment, the contents of Cd and Pb in alfalfa were far below the limit of national hygienic standard for feed, which was 30 and 1 mg/kg for Pb and Cd elements, respectively, but the content of Cr in alfalfa was close to the limit (Fig. 3a). There were no limit values for Cu and Zn in the national standard.

The calculated enrichment factors of heavy metals in alfalfa were shown in Fig. 3c and d. The EF of studied heavy metals in field experiment (Fig. 3c) decreased in the order of Cd (on average 0.90) > Cu (0.33) > Zn (0.29) > Cr (0.04) > Pb (0.01).

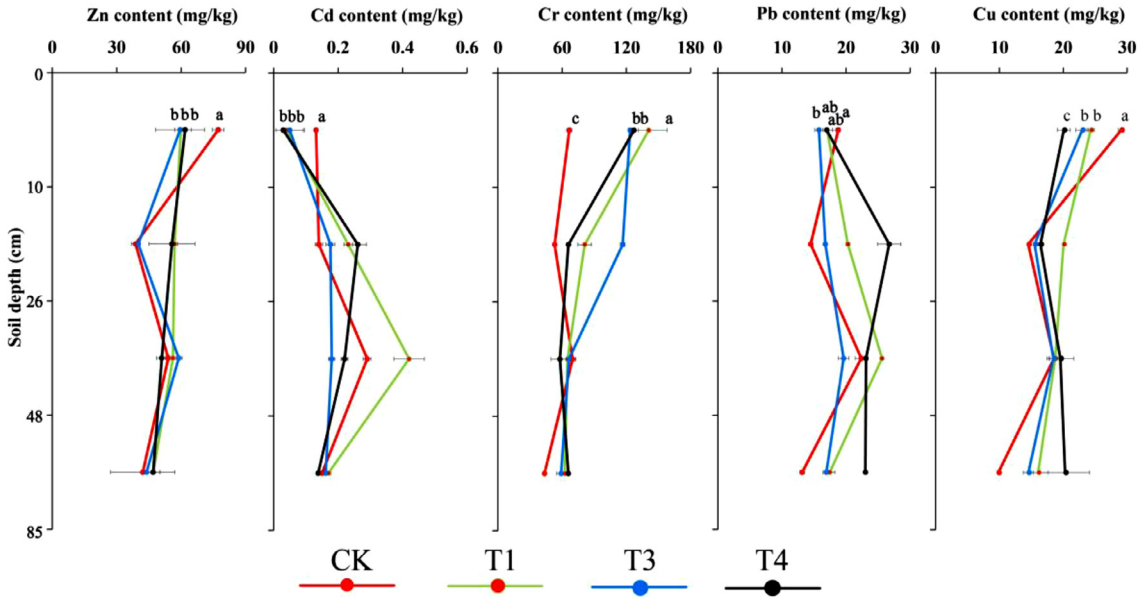

Fig. 2 – Effects of desulfurized steel slag on the content of heavy metals in saline-sodic soil profile. Different letters on the bar indicate significant differences ($p < 0.05$) between the topsoil (0–10 cm) of different treatments (CK, T1, T3, T4) according to the Duncan's test.

The EF of Cr in alfalfa in the pot experiment ranged from 0.003 to 0.01 (Fig. 3d). The DS5 and DS10 alone and combined with FA significantly increased the EF of Cr relative to the DS1 and DS1+FA treatments. However, there was no significant difference between DS alone and DS combined with FA application ($p > 0.05$).

2.4. *Bioavailable heavy metals in soils*

The concentrations and percentage of $CaCl_2$ and EDTA-extractable heavy metals in soils are presented in Appendix A Fig. S3 and Fig. 4, respectively. The concentrations of $CaCl_2$-extractable Cd and Pb, as well as EDTA-extractable Cd, were below the detection limit (0.071 μg/L for Cd and 0.296 μg/L for Pb, respectively) of ICP-MS. Except for Cu, the concentrations of EDTA-extractable Cr, Pb, and Zn were higher than those of $CaCl_2$-extractable metals (Appendix A Fig. S3). The maximum percentage of EDTA extraction was observed for Pb (on average 15.1%), followed by Zn (1.8%), Cr (0.4%), and Cu (0.3%) (Fig. 4c and d). However, the percentage of $CaCl_2$ extraction for Cr, Cu, and Zn (Fig. 4a and b) were low to below 1%, indicating that these metals had very low bioavailability. Whether DS has applied alone or combined with FA, the EDTA-extractable Cr percentage decreased with the increase of DS application dosage. However, there was no significant difference ($p < 0.05$) between DS application alone and combined with FA. Field experiment (Fig. 4c) showed that the application of DS had no significant effect on the percentage of EDTA extraction for Cu, while DS significantly decreased the percentage of EDTA extraction for Cr and Pb. The profile distribution of EDTA-extractable metals exhibited different patterns for different heavy metals (Appendix A Fig. S4). For Cd and Cu, the topsoil had a negligible extraction percentage, while subsoil had higher values (about 5%-10%). The Cr in the DS-treated soil had a very low percentage (<0.2%) in the whole soil profile. The percentage of EDTA extraction for Cd, Cu, and Pb were much higher than those of Cr and Zn.

2.5. *Chemical fractions of heavy metals in soil*

The results of sequential analyses on soil heavy metals in pot and field experiments were presented in Fig. 5. However, the concentrations of Cd for oxidizable and reducible fractions in soil were lower than the detect limit, the percentage of Cd fraction was not included in Fig. 5. The four heavy metals in saline-sodic soil existed mostly in residual form (F4), followed by reducible (F2), exchangeable (F1), and oxidizable forms (F3). The percentage of various chemical fraction for Pb in the saline-sodic soil without any amendments decreased with the following order: F4 (53.5%) > F2 (40.4%) > F3 (3.4%) > F1 (2.7%), Cr followed the decreasing order of F4 (84.8%) > F3 (10.8%) > F2 (4.0%) > F1 (0.4%), Zn followed the decreasing order of F4 (85.6%) > F2 (6.5%) > F1 (4.3%) > F3 (3.6%), and Cu had the following order of F4 (74.1%) > F2 (20.4%) > F3 (2.8%) > F1 (2.7%).

The pot and field experiments indicated that the application of DS and FA altered the chemical forms of heavy metals in soils. The weak acid extraction state of Cr in the BCR procedure was almost negligible (Fig. 5). Only Cu, Zn, and Pb had detectable exchangeable forms, accounting for 3.6%, 4.0%, and 1.9% of total metal in soils, respectively. The Cr, Cu, and Zn had a higher oxidizable fraction, which was 9.9%, 4.7%, and 3.1% of total metal content, respectively. Cd occurs mostly in the residual fractions, accounting for 94.5% of total Cd. The residual fraction of Cr, Cu, Zn, and Pb were 82.7%, 72.5%, 83.9%, and 62.2%, respectively. The application of DS decreased the percentage of exchangeable and reducible fractions of heavy metals while increasing the percentage of the residual fraction of heavy metals. Compared to the CK, the DS groups had an ele-

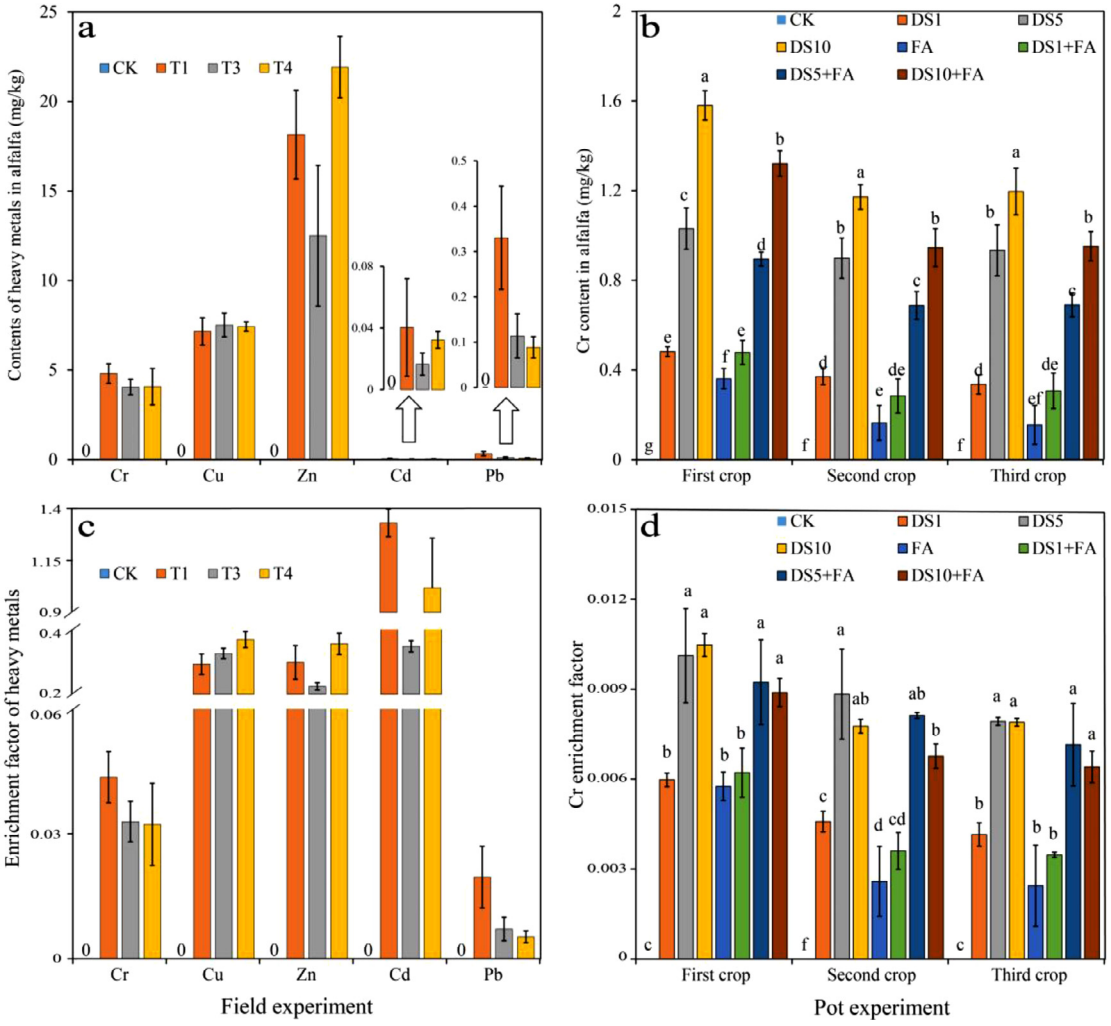

Fig. 3 – Effects of desulfurized steel slag on the content (a and b) and enrichment factor (c and d) of heavy metals in alfalfa. Different letters on the bar indicate significant differences ($p < 0.05$) according to the Duncan's test. CK, control treatment; DS1, 1% DS application; DS5, 5% DS application; DS10, 10% DS application; FA, 1% fulvic acid application; DS1+FA, 1% DS+1% fulvic acid application; DS5+FA, 5% DS+1% fulvic acid application; DS10+FA, 10% DS+1% fulvic acid application.

vated percentage of easily reducible Cu (Fig. 5). The largest increase was found in the DS10 treatment, accounting for 21.6% of total Cu. In the case of Cd, the reducible fraction was almost zero in the DS and FA-treated soils. Our previous study found that the application of DS significantly increased soil organic content (Ying et al., 2021). The elevated organic matter could contribute to increased oxidizable fraction Cu. The percentage of reducible and oxidizable Cu increased with the increased amount of DS application, while the percentage of residual form decreased. The proportion of exchangeable, reducible, and oxidizable forms of Pb decreased with the increased amount of DS application, while the proportion of residue state increased. After DS was combined with FA, the proportion of each form was almost consistent with those of DS alone.

The addition of DS significantly increased the content of Cr in soil (Table 1), so it is necessary to analyze the effect of DS improvement from the change of content of chromium. The addition of DS resulted in the change of the proportion and absolute amount of Cr in each form. In the pot experiment, the DS combined with FA treatments resulted in the partial conversion of Cr from the exchangeable or reducible form to the oxidizable or residual form than the DS alone treatments. The greatest decrease in the acid-extractable fraction (by 0.13%) was observed in the DS10+FA treatment compared with CK treatment (Fig. 5), followed by DS5+FA (by 0.1%) and DS1+FA (by 0.05%). Therefore, it can be inferred that the application of FA combined with DS can further reduce the potential toxicity of Cr, Cd, Cu, and Pb.

The results of both solution and BCR sequential extraction procedures confirmed that the application of DS decreased the bioavailability of heavy metals (Cd, Cu, and Pb) in soil. The reduction in the bioavailability of heavy metals in the soil would further reduce the accumulation of heavy metals in alfalfa. The correlation analyses among Cr content in alfalfa and soil

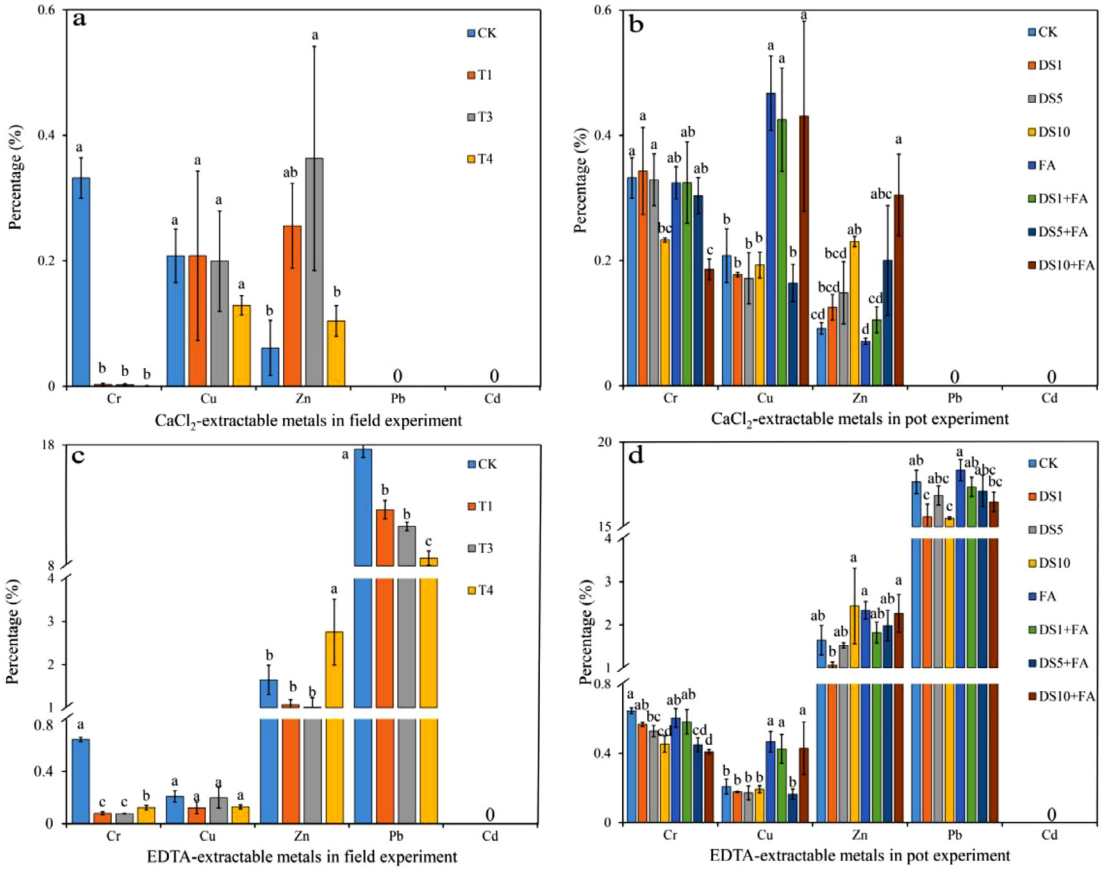

Fig. 4 – Effects of desulfurized steel slag on the percentage of $CaCl_2$ (a and b) and EDTA (c and d) extractable heavy metals in pot and field experiment (topsoil). Different letters on the bar indicate significant differences ($p < 0.05$) according to the Duncan's test.

bioavailable Cr in pot experiment were shown in Fig. 6. The Cr content in alfalfa showed significantly positive correlations with total ($R^2 = 0.876$, $p < 0.01$), $CaCl_2$- ($R^2 = 0.478$, $p < 0.01$) and EDTA-extractable Cr ($R^2 = 0.825$, $p < 0.01$) and the sum of exchangeable, reducible, and oxidizable Cr ($R^2 = 0.848$, $p < 0.01$) in soil.

3. Discussion

3.1. *Environmental risk of heavy metals in DS on soil-alfalfa system*

Our pot and field experiments indicated that the contents of studied heavy metals in the agricultural land in China (MEE and SAMR, 2018). The content of heavy metals in alfalfa grown on DS-amended soils can directly reflect the potential toxicity of heavy metals and the safety of amendment application. In the field experiment with a large DS dosage, the contents of heavy metals in growing plants met the relevant national standard. Our results confirmed that the application of DS to improve saline-sodic soil would not pollute the soil. In addition, the application rate of DS is one of the critical factors to predict the accumulation of pollutants in soil and the risk of environmental safety. If a large amount of DS has been applied to the soil for a long time, the heavy metals carried by DS may gradually accumulate in the soil over time and cause secondary pollution. According to the chromium content in DS (631 mg/kg), 571 tons of DS each needs to be applied to the soil and then the content of Cr in soil increases from background value (90 mg/kg) to pollution value (250 mg/kg) when pH > 7.5 and without leaching and migration of heavy metals (MEE and SAMR, 2018). If the input of DS is 22.5 tons/(ha•year), with no leaching or movement of heavy metals, the accumulation of Cr, Zn, Cu, and Pb from uncontaminated background soil to contaminated soils in pH > 7.5 would need 25, 870, 1083, and 4500 years, respectively. Under other pH values, the year of pollution caused by DS input was shown in Appendix A Table S3. Actually, the annual dosage of iron and steel slags used as a soil amendment in China was about 2–50 tons/ha (Wang et al., 2021a). Therefore, it could be concluded that the accumulation of heavy metals in soils caused by DS addition at low dosage each year could not exceed the screening value of soil pollution risk of agricultural land in China. However, it was predictable that the application of DS in a long-term and larger dosage might result in pollution risks of heavy metals Cr. In order to reduce the input of exogenous Cr from the addition of DS, the application rate of DS needs to be controlled at a certain level.

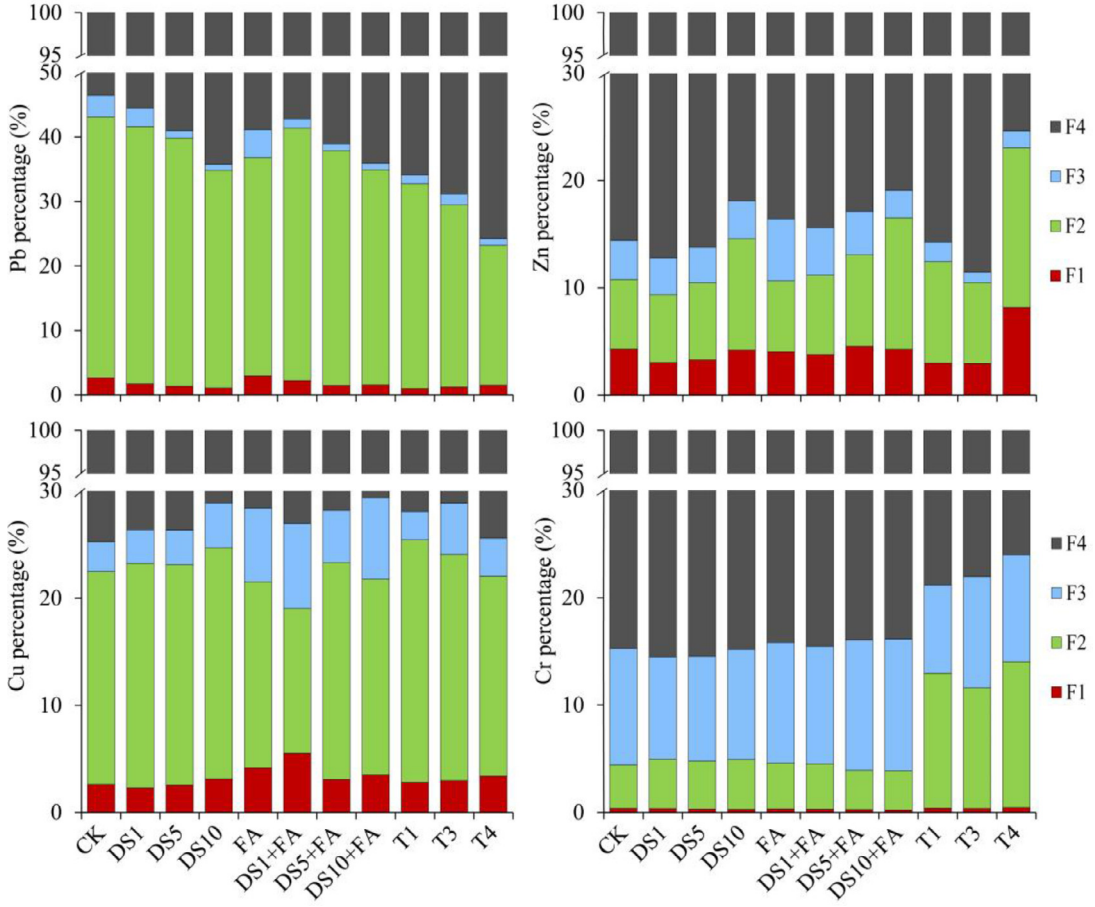

Fig. 5 – Effect of desulfurized steel slag on chemical fractions of heavy metals in pot and field experiments. The fractionation of heavy metals in the soil was performed using the BCR sequential extraction procedure. The heavy metals in soil were fractionated into the exchangeable (F1), Fe-Mn oxides-bound or easily reducible (F2), organically bound fraction or oxidizable (F3) and residual fractions (F4), respectively.

3.2. *Mechanisms for environmental safety of heavy metals in DS*

An important mechanism for the environmental safety of DS as soil amendment was the bioavailability of heavy metals in soil. The bioavailable form of heavy metals in soil can be used as a useful indicator of soil pollution (Kim et al., 2015). In fact, heavy metals of DS existing in hydroxide, silicates, and ferrite precipitates resulted in deficient mobility and release of heavy metals in DS itself. The specific extracted-Cd experiment in DS also exhibited that release of Cr from the slag could be ignored. Our results agreed with those already reported in the literature (Dong et al., 2021; Ragipani et al., 2020; Singh et al., 2021). Theoretically, it is a non-toxic material. The studies in Japan and Europe also showed that the heavy metal elements in steel slag (such as Cr) are not quickly and easily released into the environment (Ragipani et al., 2020; Singh et al., 2021). In addition, slag exposure to natural environments leads to the formation of carbonate phases, which effectively reduce the leaching and release of heavy metals into the soil (Li et al., 2022; Santos et al., 2012). The formation of heavy metals precipitates bound within stable carbonate phases effectively prevents the plant adsorption of heavy metals. Therefore, the existence of various iron oxides in DS can effectively reduce the toxicity of Cr. These potential mechanisms for stable heavy metals in DS could explain the very low release of heavy metals into soil.

Another mechanism for the environmental safety of DS as soil amendment was that application of DS decreased the mobility of heavy metals (Cu, Pb, and Zn), and the bioavailable forms (F1) transformed into relatively stable forms, such as residual states (Fig. 5). Generally, the exchangeable part of heavy metals is retained on the colloidal surface by electrostatic force, which represents the metal most easily absorbed and utilized by organisms (Filgueiras et al., 2002; Rao et al., 2008; Zhang et al., 2014). The reducible part of heavy metals is mainly bonded around iron-manganese oxides, which are unstable in an anaerobic or anoxic environment. Hence, the reducible fraction of heavy metals could be considered as potentially bioavailable metals. The oxidizable state of heavy metals is regarded as the metal in stable and high molecular weight humus or appears as oxidizable minerals, which is usually not easy to be bioavailable (Mäkelä et al., 2012). The residual state of heavy metals is generally retained in the crystal structure. Therefore, the residual state of heavy metals is very stable under natural conditions, and the bioavailability can be ignored.

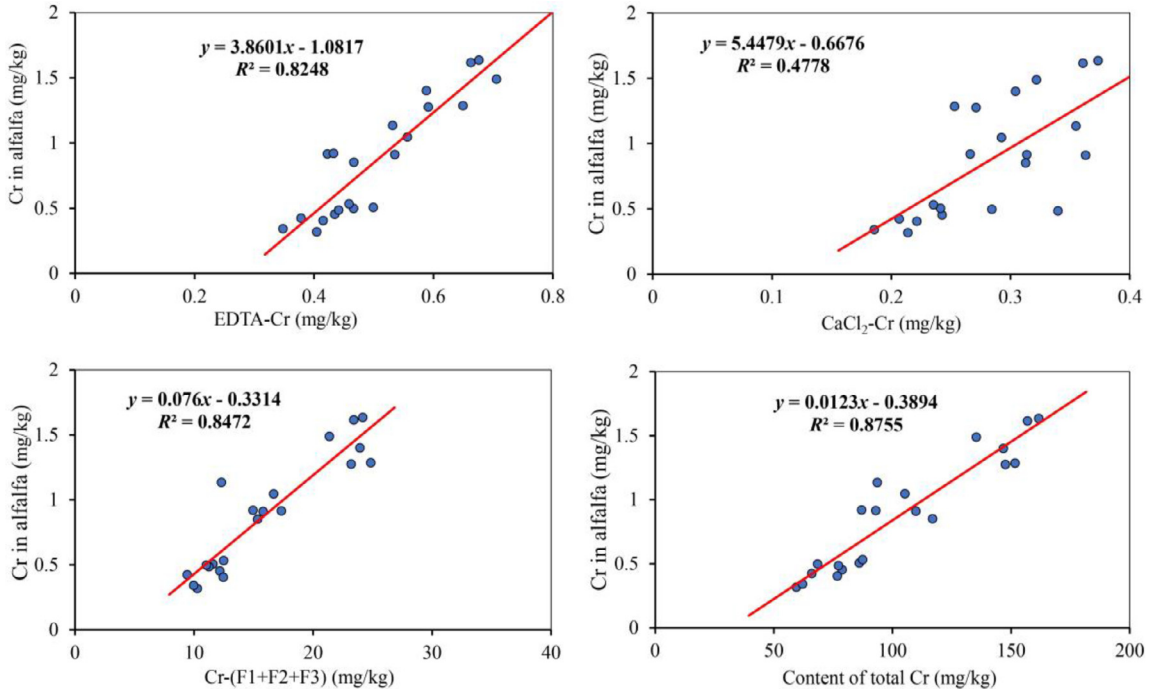

Fig. 6 – Correlations between Cr content in alfalfa and EDTA-extractable Cr, $CaCl_2$- Cr, sum of exchangeable (F1), reducible (F2) and oxidizable (F3) Cr, and total Cr in pot experiment.

In addition, it is found that fulvic acid in soil is mainly adsorbed on the surface of iron oxide, and fulvic acid could form stable complexes with heavy metals (especially Cr), which further reduced the risk of heavy metals in soil (Li et al., 2022). In our experiments, the application of DS combined with FA showed a more significant effect in enhancing plant growth (Appendix A Fig. S2) and decreasing Cr accumulation in alfalfa compared to the control and DS alone treatments (Fig. 3b and d). This could explain why the activities of Cr decreased after FA treatment in this pot experiment. This suggested that DS in combination with FA addition may be a potential method for not only reducing the accumulation of heavy metals but enhancing alfalfa safety production.

3.3. *The feasibility of the use of DS in agriculture*

In order to judge the feasibility of DS in agricultural use, the influence of DS utilization on the content of heavy metals in the soil and crop should be addressed carefully. Based on the results, the application of DS at a large dosage to soil may lead to excessive accumulation of Cr in alfalfa, endangering feed safety. Furthermore, accurate calculations indicated that when the DS application amount was 0.9 tons/ha, the Cr content in DS-amended soil increased by less than 1% compared with the control group. Therefore, when DS is applied as a soil conditioner, the risk is expected to be low, especially at a reasonable amount.

Secondly, whether the DS can be applied in agriculture depends on the particular concern Cr. The range of chromium content in steel slag mainly depends on the composition of raw materials used in steel production (Wang et al., 2021a). In soil, chromium primarily exists in positive trivalent and positive hexavalent (Park, 2020). A small amount of trivalent chromium is an essential nutrient, while hexavalent chromium is highly toxic (Hamilton, 2020). Thus, it can be inferred that the transformation of Cr form and valence state would cause more significant environmental risks. In addition, there are a lot of electron acceptors in free oxides and active oxides in steel slag (Navarro et al., 2010; Proctor et al., 2000; Wang et al., 2021a). The existence of low valent iron in steel slag, such as Fe^0/Fe^{2+} can effectively reduce hexavalent chromium to trivalent chromium, and the dissolved iron ions and chromium ions further exist in the form of mixed oxides or hydroxides (Tang et al., 2014). Moreover, because DS is rich in calcium oxide and magnesium oxide, it will also be a good option for CO_2 storage absorbent. The DS-amended soil could sequestrate more carbon, thereby helping to slow down global warming (Kumar et al., 2020; Liu et al., 2021; Ragipani et al., 2020; Reddy et al., 2019). Therefore, the DS could be viewed as a potential feedstock for CO_2 sequestration.

The application of DS in saline-sodic soils could effectively ameliorate poor soil properties and promote alfalfa growth (Ying et al., 2021). The DS contained rich calcium sulfate, which is very effective in improving the pH of saline-sodic soil. The accumulation of heavy metals was within the screening value of soil pollution risk in agricultural land. Therefore, the characteristics of DS are very suitable for use in ameliorating saline-sodic soil. The application of DS in the improvement of saline-sodic land can reduce the load of steel slag on the environment simultaneously, so it has great potential in agriculture. The use of DS could mitigate the exploitation of natural resources, cost, and energy, control air pollutants, and amelio-

rate soil, which provided a practice possibility for win-win-win strategy, flue gas desulfurization, saline-sodic soil improvement, and utilization of solid wastes. However, the content of heavy metals in DS from different steel slags varies greatly, it is necessary to study the risk of heavy metal pollution in the agricultural application of DS from different sources.

4. Conclusions

Both pot and field experimental results confirmed that the application of desulphurized steel slag (DS) in agriculture is safe considering the heavy metal (Cr, Cd, Cu, Pb, and Zn) contents in the soil and plants grown on it. Due to the higher content of Cr in DS, the content of Cr in the DS-amended soil increased significantly. The heavy metals (Cu, Pb, and Zn) in DS-amended soils could be transformed from an easily mobile state to insoluble residual form, thereby reducing the bioavailability and mobility of heavy metals in soils. The combined application of DS and FA significantly enhanced the biomass of alfalfa and decreased Cr accumulation in the alfalfa. It may be a possible way for the safe production of alfalfa in saline-sodic soil. This study confirmed that the direct application of DS in agriculture is a safe and feasible technology, which had relatively little pollution risk of Cr to the soil-crop system.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A Supplementary data

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.jes.2022.05.010.

REFERENCES

Annunziata Branca, T., Pistocchi, C., Colla, V., Ragaglini, G., Amato, A., Tozzini, C., et al., 2014. Investigation of (BOF) converter slag use for agriculture in Europe. Metall. Res. Technol. 111, 155–167.

Cha, W., Kim, J., Choi, J.H., 2006. Evaluation of steel slag for organic and inorganic removals in soil aquifer treatment. Water Res. 40, 1034–1042.

Chang, S., Zhuo, J., Meng, S., Qin, S., Yao, Q., 2016. Clean coal technologies in China: current status and future perspectives. Engineering 2, 447–459.

Chen, L., Dick, W.A., Nelson, S., 2001. Flue gas desulfurization by-products additions to acid soil: alfalfa productivity and environmental quality. Environ. Pollut. 114, 161–168.

Chen, Q., Wang, S.J., Li, Y., Zhang, N., Zhao, B., Zhuo, Y.Q., et al., 2015. Influences of flue gas desulfurization gypsum as amendment on heavy metal distributions in reclaimed sodic soils. Environ. Eng. Sci. 32, 1–9.

Clark, R., Ritchey, K., Baligar, V., 2001. Benefits and constraints for use of FGD products on agricultural land. Fuel 80, 821–828.

86/278/EEC, 1986. Council of the European communities council directive of 12 June 1986 on the protection of the environment, and in particular of the soil, when sewage sludge is used in agriculture. Off. J. Eur. Communities 181, 6–12.

Cui, L., Liu, M., Yuan, X., Wang, Q., Ma, Q., Wang, P., et al., 2021. Environmental and economic impact assessment of three sintering flue gas treatment technologies in the iron and steel industry. J. Clean. Prod. 311, 127703.

Dong, Q., Wang, G., Chen, X., Tan, J., Gu, X., 2021. Recycling of steel slag aggregate in portland cement concrete: an overview. J. Clean. Prod. 282, 124447.

Filgueiras, A.V., Lavilla, I., Bendicho, C., 2002. Chemical sequential extraction for metal partitioning in environmental solid samples. J. Environ. Monit. 4 (6), 823–857.

Fisher, L.V., Barron, A.R., 2019. The recycling and reuse of steel making slags – a review. Res. Conserv. Recycl. 146, 244–255.

Francisca, F.M., Glatstein, D.A., 2020. Environmental application of basic oxygen furnace slag for the removal of heavy metals from leachates. J. Hazard. Mater. 384, 121294.

Fuentes, A., Lloréns, M., Sáez, J., Soler, A., Aguilar, M.I., Ortuno, J.F., et al., 2004. Simple and sequential extractions of heavy metals from different sewage sludges. Chemosphere 54, 1039–1047.

Guo, J.L., Bao, Y.P., Wang, M., 2018. Steel slag in China: treatment, recycling, and management. Waste Manag. 78, 318–330.

Hamilton, E.M., Young, S.D., Bailey, E.H., Humphrey, O.S., Watts, M.J., 2020. Assessment of chromium species dynamics in root solutions using isotope tracers. J. Trace Elem. Med. Biol. 61, 126514.

He, H.D., Tam, N.F.Y., Yao, A.J., Qiu, R.L., Li, W.C., Ye, Z.H., 2017. Growth and Cd uptake by rice (*Oryza sativa*) in acidic and Cd-contaminated paddy soils amended with steel slag. Chemosphere 189, 247–254.

Kambole, C., Paige-Green, P., Kupolati, W.K., Ndambuki, J.M., Adeboje, A.O., 2017. Basic oxygen furnace slag for road pavements: a review of material characteristics and performance for effective utilisation in southern Africa. Constr. Build. Mater. 148, 618–631.

Kim, R.Y., Yoon, J.K., Kim, T.S., Yang, J.E., Owens, G., Kim, K.R., 2015. Bioavailability of heavy metals in soils: definitions and practical implementation-a critical review. Environ. Geochem. Health 37, 1041–1061.

Koralegedara, N.H., Pinto, P.X., Dionysiou, D.D., Al-Abed, S.R., 2019. Recent advances in flue gas desulfurization gypsum processes and applications – a review. J. Environ. Manag. 251, 109572.

Kumar, S.S., Kumar, A., Singh, S., Malyan, S.K., Baram, S., Sharma, J., et al., 2020. Industrial wastes: fly ash, steel slag and phosphogypsum- potential candidates to mitigate greenhouse gas emissions from paddy fields. Chemosphere 241, 124824.

Li, L., Ling, T.C., Pan, S.Y., 2022. Environmental benefit assessment of steel slag utilization and carbonation: a systematic review. Sci. Total Environ. 806, 150280.

Liu, S., Liu, W., Jiao, F., Qin, W., Yang, C., 2021. Production and resource utilization of flue gas desulfurized gypsum in China – a review. Environ. Pollut. 288, 117799.

Lu, R., 2000. Soil and Agro-Chemical Analysis Methods. Agricultural Science and Technology Press, Beijing.

Mäkelä, M., Watkins, G., Pöykiö, R., Nurmesniemi, H., Dahl, O., 2012. Utilization of steel, pulp and paper industry solid residues in forest soil amendment: relevant physicochemical properties and heavy metal availability. J. Hazard. Mater. 207-208, 21–27.

Matveeva, V., Lytaeva, T., Danilov, A., 2018. Application of steel-smelting slags as material for reclamation of degraded lands. J. Ecol. Eng. 19, 97–103.

Meng, Z.H., Wang, C.Y., Wang, X.R., Chen, Y., Li, H.Q., 2018. Simultaneous removal of SO_2 and NOx from coal-fired flue gas using steel slag slurry. Energy Fuels 32, 2028–2036.

Meng, Z.H., Wang, C.Y., Wang, X.R., Chen, Y., Wu, W.F., Li, H.Q., 2019. Simultaneous removal of SO_2 and NOx from flue gas using $(NH_4)_2S_2O_3$/steel slag slurry combined with ozone oxidation. Fuel 255, 115720.

GB 15618-2018 (Soil environmental quality: Risk control standard for soil contamination of agricultural land), 2018. Ministry of Ecology and Environment and State Administration for Market Regulation of the People's Republic of China (MEE and SAMR). China Environmental Science Press, Beijing (accessed 28 October 2020).

Navarro, C., Díaz, M., Villa-García, M.A., 2010. Physico-chemical characterization of steel slag. Study of its behavior under simulated environmental conditions. Environ. Sci. Technol. 44, 5383.

Nurmesniemi, H., Pöykiö, R., Watkins, G., Dahl, O., 2010. Total and extractable heavy metal, phosphorous and sulfur concentrations in slaker grits from the causticizing process of a pulp mill for use as a soil amendment. Chem. Speciation Bioavail. 22, 87–97.

Park, J.H., 2020. Contrasting effects of Cr(III) and Cr(VI) on lettuce grown in hydroponics and soil: chromium and manganese speciation. Environ. Pollut. 266 (Pt 2), 115073.

Piatak, N.M., Seal II, R.R., Hoppe, D.A., Green, C.J., Buszka, P.M., 2019. Geochemical characterization of iron and steel slag and its potential to remove phosphate and neutralize acid. Minerals 9, 468.

Pistocchi, C., Ragaglini, G., Colla, V., Branca, TA., Tozzini, C., Romaniello, L., 2017. Exchangeable sodium percentage decrease in saline sodic soil after basic oxygen furnace slag application in a lysimeter trial. J. Environ. Manag. 203, 896–906.

Proctor, D.M., Fehling, K.A., Shay, E.C., Wittenborn, J.L., Green, J.J., Avent, C., et al., 2000. Physical and chemical characteristics of blast furnace, basic oxygen furnace, and electric arc furnace steel industry slags. Environ. Sci. Technol. 34, 1576–1582.

Ragipani, R., Bhattacharya, S., Akkihebbal, S.K., 2020. Understanding dissolution characteristics of steel slag for resource recovery. Waste Manag. 117, 179–187.

Rao, C.R.M., Sahuquillo, A., Lopez Sanchez, J.F., 2008. A review of the different methods applied in environmental geochemistry for single and sequential extraction of trace elements in soils and related materials. Water Air Soil Pollut. 189, 291–333.

Reddy, K.R., Gopakumar, A., Chetri, J.K., 2019. Critical review of applications of iron and steel slags for carbon sequestration and environmental remediation. Rev. Environ. Sci. Bio 18, 127–152.

Santos, R.M., Ling, D., Sarvaramini, A., Guo, M.X., Elsen, J., Larachi, F., et al., 2012. Stabilization of basic oxygen furnace slag by hot-stage carbonation treatment. Chem. Eng. J. 203, 239–250.

Scott, I.S.P.C., Penn, C.J., 2021. Estimating the variability of steel slag properties and their influence in phosphorus removal ability. Chemosphere 276, 130205.

Singh, S.K., Vashistha, P., Chandra, R., Rai, A.K., 2021. Study on leaching of electric arc furnace (EAF) slag for its sustainable applications as construction material. Process Saf. Environ. Prot. 148, 1315–1326.

Smeda, A., Zyrcniki, W., 2002. Application of sequential extraction and the ICP-AES method for study of the partitioning of metals in fly ashes. Microchem. J. 72, 9–16.

Srivastava, R.K., Jozewicz, W., 2001. Flue gas desulfurization: the state of the art. J. Air Waste Manag. Assoc. 51 (12), 1676–1688.

GB 4284-2018 (Control standards of pollutants in sludge for agricultural use), 2018. State Administration for Market Regulation of the People's Republic of China and National Standardization Management Committee (SAMR and NSMC). China Environmental Science Press, Beijing.

Tang, W.W., Zeng, G.M., Gong, J.L., Liang, J., Xu, P., Zhang, C., et al., 2014. Impact of humic/fulvic acid on the removal of heavy metals from aqueous solutions using nanomaterials: a review. Sci. Total Environ. 468, 1014–1027.

Wang, X., Cai, Q.S., 2006. Steel slag as an iron fertilizer for corn growth and soil improvement in a pot experiment. Pedosphere 16, 519–524.

Wang, W., Sardans, J., Lai, D.Y.F., Wang, C., Zeng, C., Tong, C., et al., 2015. Effects of steel slag application on greenhouse gas emissions and crop yield over multiple growing seasons in a subtropical paddy field in China. Field Crops Res. 171, 146–156.

Wang, S.J., Chen, Q., Li, Y., Zhuo, Y.Q., Xu, L.Z., 2017. Research on saline-alkali soil amelioration with FGD gypsum. Res. Conserv. Recycl. 121, 82–92.

Wang, X.B., Li, X.Y., Yan, X., Tu, C., Yu, Z.G., 2021a. Environmental risks for application of iron and steel slags in soils in China: a review. Pedosphere 31, 28–42.

Wang, Y., Wang, Z., Liang, F., Jing, X., Feng, W., 2021b. Application of flue gas desulfurization gypsum improves multiple functions of saline-sodic soils across China. Chemosphere 277, 130345.

Wang, T., Lou, Y.B., Jiang, S.C., Wang, J.W., Zhang, Y.S., Pan, W.P., 2022. Distribution characteristics and environmental risk assessment of trace elements in desulfurization sludge from coal-fired power plants. Fuel 314, 122771.

Wen, T.T., Yang, L.Y., Dang, C.Y., Miki, T., Bai, H., Nagasaka, T., 2020. Effect of basic oxygen furnace slag on succession of the bacterial community and immobilization of various metal ions in acidic contaminated mine soil. J. Hazard. Mater. 388, 121784.

Yang, B.M., Lai, W.L., Chang, Y.M., Liang, Y.S., Kao, C.M., 2017. Using desulfurization slag as the aquacultural amendment for fish pond water quality improvement: Mechanisms and effectiveness studies. J. Clean. Prod. 143, 1313–1326.

Yi, H., Xu, G.P., Cheng, H.G., Wang, J.S., Wan, Y.F., Chen, H., 2012. An overview of utilization of steel slag. Procedia Environ. Sci. 16, 791–801.

Ying, Y.Q., Lu, S.G., Shi, H.X., Shi, Y.Z., Shi, Y.N., Rao, W.J., et al., 2021. Flue gas desulfurization (FGD) steel slag ameliorates salinity, sodicity, and adverse physical properties of saline-sodic soil of middle Yellow River, China. Environ. Sci. Pollut. Res. 28, 36765–36774.

Zhang, C., Yu, Z.G., Zeng, G.M., Jiang, M.Y., Zhong, Z.C., Fang, Z., et al., 2014. Effects of sediment geochemical properties on heavy metal bioavailability. Environ. Int. 73, 270–281.

Zhang, X., Chen, J., Jiang, J., Li, J., Tyagi, R.D., Surampalli, R.Y., 2020. The potential utilization of slag generated from iron and steelmaking industries: a review. Environ. Geochem. Health 42, 1321–1334.

Zhao, Y., Wang, S., Li, Y., Zhuo, Y., Liu, J., 2019. Sustainable effects of gypsum from desulphurization of flue gas on the reclamation of sodic soil after 17 years. Eur. J. Soil Sci. 70, 1082–1097.